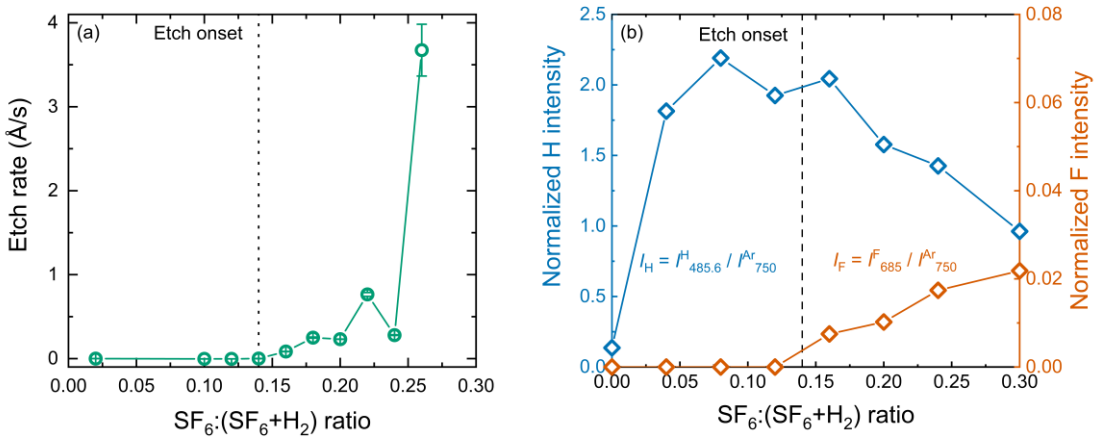

Figure 1: Characterization of the fluorination half-cycle. (a) Etch rate of MoS_2 by exposure to the SF_6/H_2 plasma as function of the ratio of the plasma flow rates. Below the etch onset the MoS_2 film is not etched, but only modified. (b) H and F radical intensities normalized to the Ar intensity as measured by optical emission spectroscopy. Below the etch onset no F radicals are observed, showing that enough H_2 was added to scavenge the F radicals responsible for continuous etching.

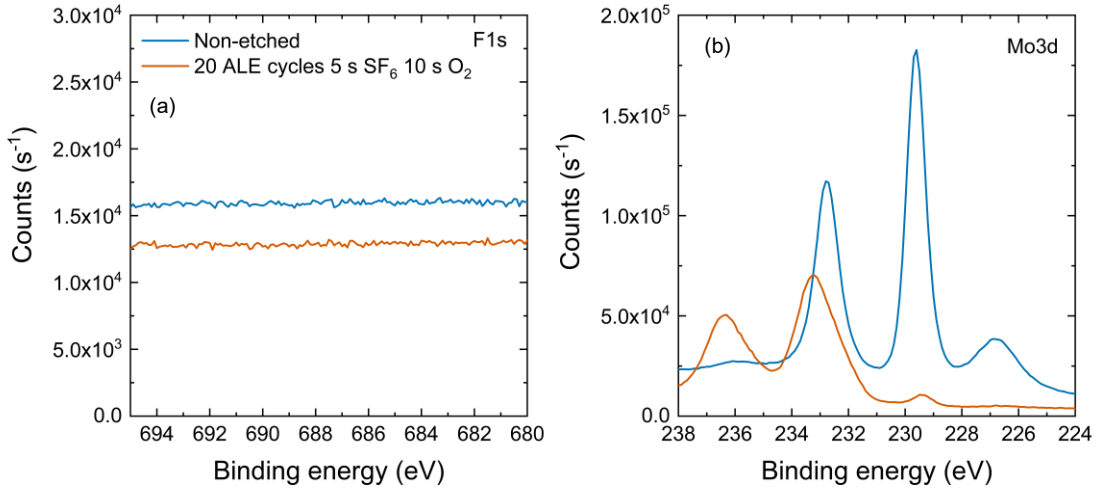

Figure 2: XPS spectra of (a) F1s and (b) Mo3d of non-etched film and film etched with 20 ALE cycles. No F contamination was measured. The Mo peaks show a shift to higher binding energies after ALE, indicating oxidation.

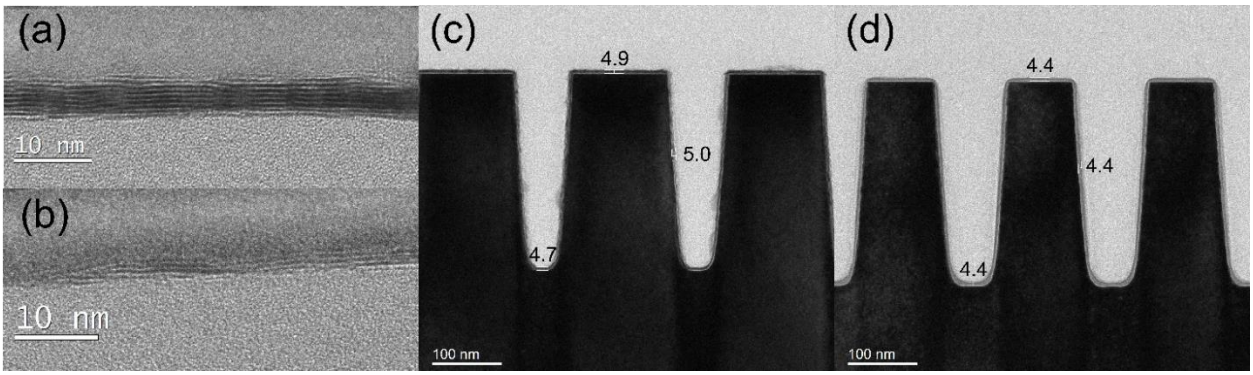

Figure 3: Cross-sectional bright field transmission electron microscopy images of MoS_2 (a,c) before, (b) after 20 ALE cycles, and (d) after 10 ALE cycles. (a,b) Films deposited on a planar substrate. Boundaries between crystalline MoS_2 grains are visible as vertical dark lines. (b) The diffuse darker layer on top of the crystalline 2D layers is assumed to be an amorphous MoO_x top layer. (c,d) Films deposited in 3D Si trenches. Thickness of the film is indicated in nm at different locations. A similar amount of MoS_2 is etched on all sides, indicating that the ALE process is isotropic. Due to the amorphous top layer seen in (b), the total thickness of the film in (c,d) does not change much as a result of the ALE process. Larger structures are visible in (c); they are assumed to be out-of-plane oriented structures (fins). In (d) the larger structures are no longer visible, suggesting that the fins are etched.